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Development of Laser Mirrors of Very High Reflectivity Using the Cavity-Attenuated Phase-Shift (CAPS) Method

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Prepared by
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Aerophysics Laboratory

5 August 1981

Prepared for
VICE PRESIDENT AND GENERAL MANAGER
LABORATORY OPERATIONS

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El Segundo, Calif. 90245

ABSTRACT

It has been possible to obtain mirrors of very high reflectivity by following the simple four-step procedure described herein. The key to success is the ability to measure the scattering and other losses of the substrates and dielectric coatings to ensure that the specifications are being met. These measurements are especially critical in the important cleaning process. The cavity-attenuated phase-shift (CAPS) method is ideally suited for performing these important measurements, permitting us to obtain mirrors with reflectivities of $R = 0.99975 \pm 0.00005$.

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I. INTRODUCTION

High-power infrared laser systems and shorter wavelength laser systems based upon electronic transitions have created a need for very-high-reflectivity laser mirrors. This requirement arises from the extremely high photon-flux densities associated with these laser devices and the much lower increases in power per transit between mirrors (gain) of the new electronic transition laser systems compared to the infrared laser systems. As a case in point, for the nitrogen fluoride system currently being studied as a potential laser candidate, the gain is so low that mirrors in excess of 99.95% reflectivity are required for an initial lasing demonstration on a laboratory scale.

We are pleased to report that, with the help of several specialist groups in the field, we have been able to produce mirrors with reflectivities of 99.975% (at a wavelength value near 0.87 μm) and that other advances in the state of the art could be achieved if there were a requirement. This accomplishment was possible because we invented a new method for making the critical scattering loss measurements associated with the mirror substrate cleaning process, i.e., the cavity-attenuated phase-shift (CAPS) method.

II. HIGH-REFLECTIVITY MIRROR DEVELOPMENT PROCEDURE

The procedure that we used to produce the high-reflectivity mirrors consists of four basic steps: (1) the procurement of high-quality quartz substrates from a suitable vendor, (2) the cleaning, measurement, and preparation of these substrates, (3) the coating process by which a low-scatter dielectric coating is applied by a qualified supplier, and (4) the measurement of the resultant mirror reflectivity. Since Steps (1) and (3) are performed by suppliers, only Steps (2) and (4) are necessary for ensuring the desired quality.

A. SUBSTRATE PROCUREMENT

The quartz substrates were obtained from General Optics. These were specified to have an RMS smoothness of ≤ 10 Å and be flat and parallel to $\leq \lambda/4$. These substrates had been cleaned by General Optics. To remove the excess grinding material and waxes used in the polishing process, substantially more cleaning was required. Figure 1 is a photograph taken of one of these General Optics quartz substrates before cleaning. A dark-field microscope at 400X magnification was used. Since the field of view is approximately 0.5 mm, the larger particles are approximately 100 μm with the particles ranging to a detection limit of about 10 μm.

B. SUBSTRATE CLEANING

In our search for a reliable and efficient cleaning procedure, we quickly discovered that each researcher had his own special technique. After some deliberation, we decided to use a "try and see" approach. We started with the special solvent mixture of 1,1,1, trichlorethane and ethanol, which was specially treated to minimize all nonvolatile residues, from Analytical Research

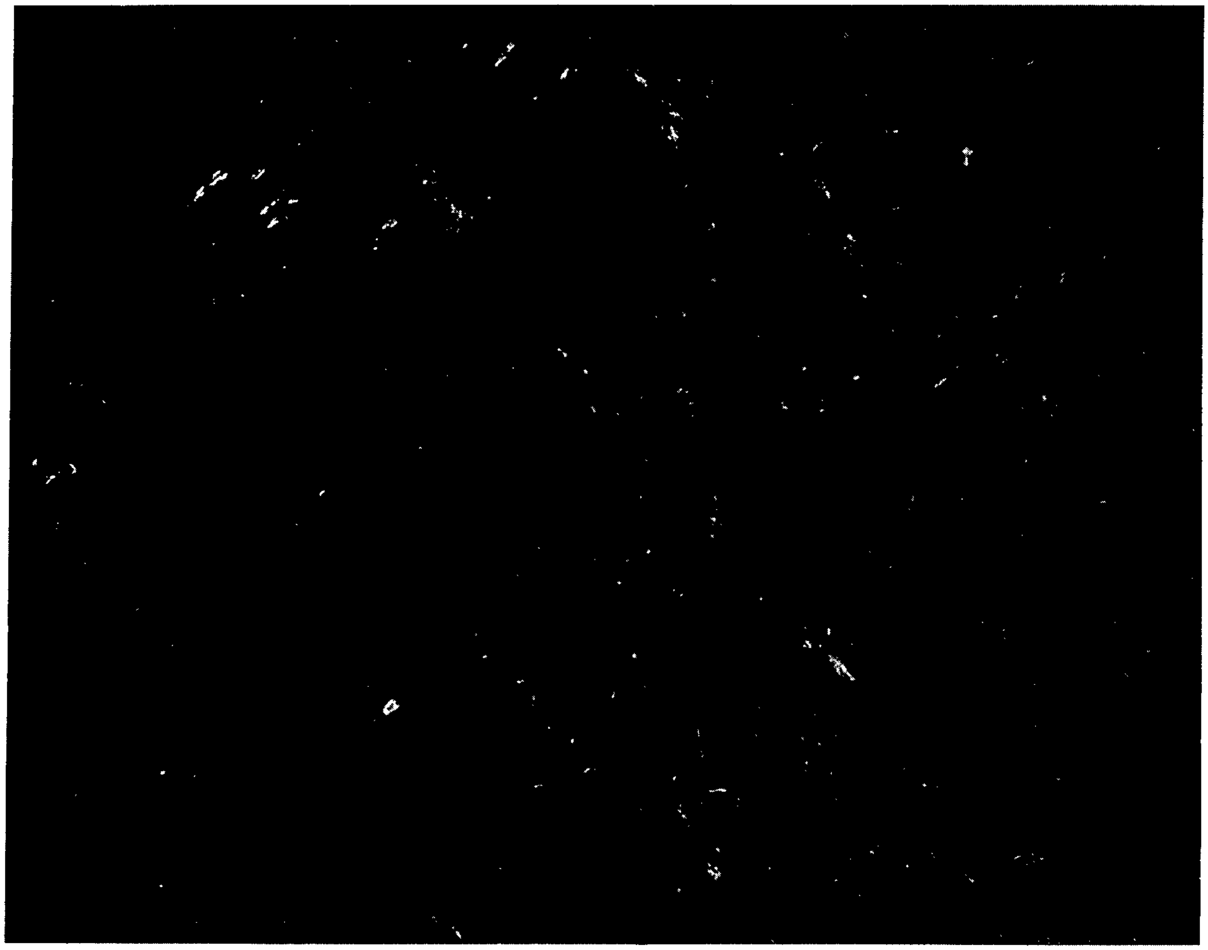

Fig. 1. Photograph of a Quartz Substrate Before Cleaning as Observed Through a Dark-Field Microscope at 400X Magnification. The field of view is 0.5 mm.

Laboratory, Inc. Figure 2 is a photograph of a quartz substrate after being cleaned with this "super solvent." The improvement is obvious. Various cleaning or rubbing techniques were attempted, and microscopic examination followed. We discovered that each technique has its own advantages, and it was difficult to decide which was the most beneficial. It was difficult to correlate the microscope pictures with a quantitative estimate of the scattering. Consequently, we decided to measure the scattering from the surfaces by means of the photon lifetime measurement technique recently developed in this laboratory.[1] Using a modification of this technique we were able to quantitatively measure the scattering and to determine the quickest, easiest, and most reliable technique for cleaning the quartz substrates. The ability to make these quantitative scattering measurements has significantly reduced the labor and equipment costs involved while substantially increasing the reliability of the cleaning procedure.

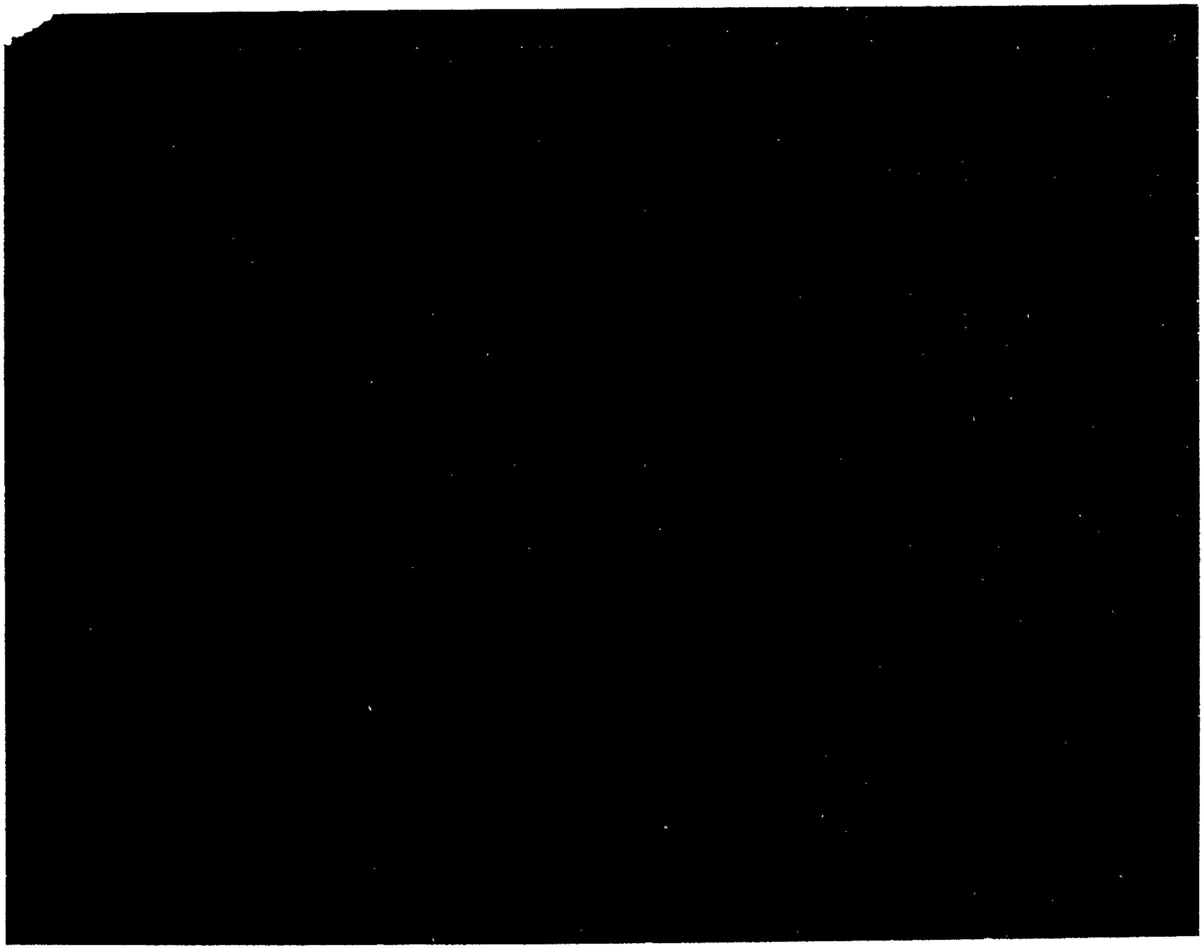

Fig. 2. Photograph of a Cleaned Quartz Substrate as Observed Through a Dark-Field Microscope at 400X Magnification. The field of view is 0.5 mm.

III. QUARTZ SUBSTRATE SCATTERING MEASUREMENTS

This technique is described in detail elsewhere.[1] A brief description is provided here for convenience. Figure 3 is a schematic of the experimental setup. An amplitude modulated light beam from a suitable source, usually a laser, is directed through one of the slightly transmitting mirrors into the aligned optical cavity comprised of the mirrors and optics to be measured. The dissipation of the light energy within the cavity is caused by absorption, scattering, or transmission at the surfaces of the cavity mirrors and at other optical components within the resonator. Each loss shortens the effective photon lifetime, which is defined as the characteristic time τ for the photon energy to be dissipated to e^{-1} (or 0.34) of its original value. This photon lifetime is determined from a phase shift in the light-beam intensity that has passed through the optical resonator.

The amplitude modulation, which is required to extract time-dependent lifetime data from a steady-state experiment, is conveniently produced by passing the continuous source beam through a piezo-optical birefringence modulator. A linearly polarized photon beam with a $\sin^2 (2\pi\ ft + \phi)$ modulation at $f = 50$ kHz is produced. The resultant phase shift α of the emerging beam is related to the photon lifetime by the simple expression, $\tan(\alpha) = 4\ \pi\ f\ \tau$. Since this lifetime corresponds to some number n of round trips that the photons make within the optical resonator $n = c\tau/2L$, where c is the speed of light, and L is the distance between the mirrors, the overall loss of the optical resonator $1 - R$ and, therefore, the product of the reflectivities $R = R_1R_2$ can be obtained from the simple formula $1 - R = 1/(n + 1)$.

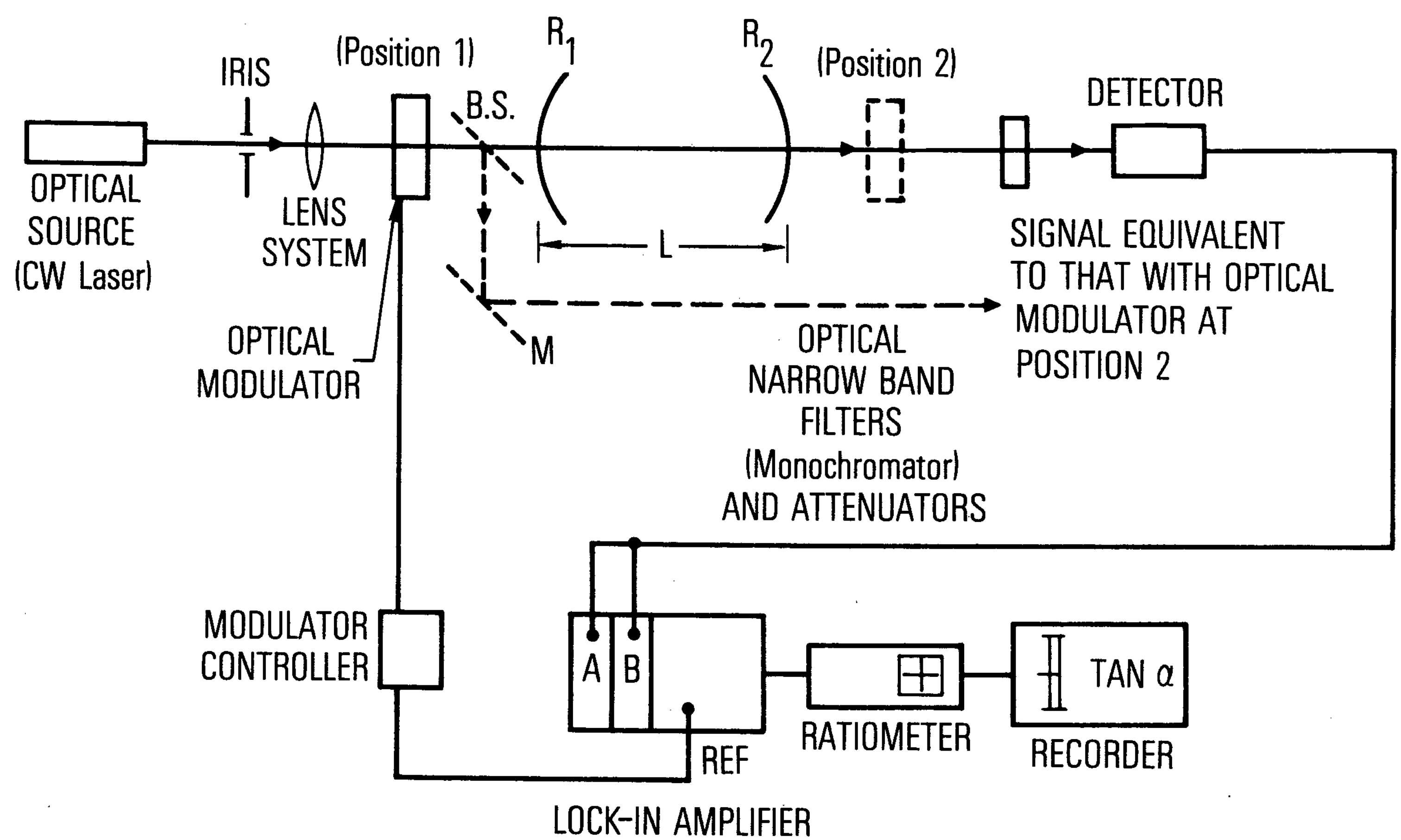

Fig. 3. Physical Layout of Optic Train and Phase Shift Measurement Equipment Used to Measure Average Lifetime of Photons in Optical Resonator

With the use of appropriate combinations of different cavity mirrors and other optical components, each of the respective losses and reflectivities can be determined. The most convenient method of measurement is to use a two-phase lock-in amplifier to measure both the sin α and cos α, which are, in turn, input into a ratiometer to obtain tan α directly. For large n's (n typically ranges from 200 to 2000), this output signal is directly proportional to the cavity lifetime, affording on-line in-situ measurements.

The two ways that the quartz substrate can be placed into the optical cavity to minimize reflectance losses, i.e., at Brewster's angle and perpendicular to the cavity axis, are shown in Fig. 4. At the Brewster's angle, by definition, the reflectance loss for the perpendicular polarized light, which is what is being introduced into the cavity, is theoretically very close to zero. This assumption has been used by other investigators as a means of calibrating their mirror reflectivity measurement methods.[2] However, we quickly discovered that the substrate placed at the perpendicular angle showed considerably less attenuation of the phase shift and, therefore, less loss than when the same substrate was placed at Brewster's angle. Although the reflectance off the surfaces of the substrate at normal angle is much larger, approximately 4%, it is reflected right back at the mirror and therefore does not represent a loss. However, when the substrate is placed at the Brewster's angle, the light which is plane polarized perpendicular to the plane of incidence is reflected out of the cavity while the light polarized parallel to the plane of incidence is not. Consequently, any rotation of the plane of polarization within the cavity, such as through the birefringence of the quartz substrate itself, results in a much higher loss than would have been

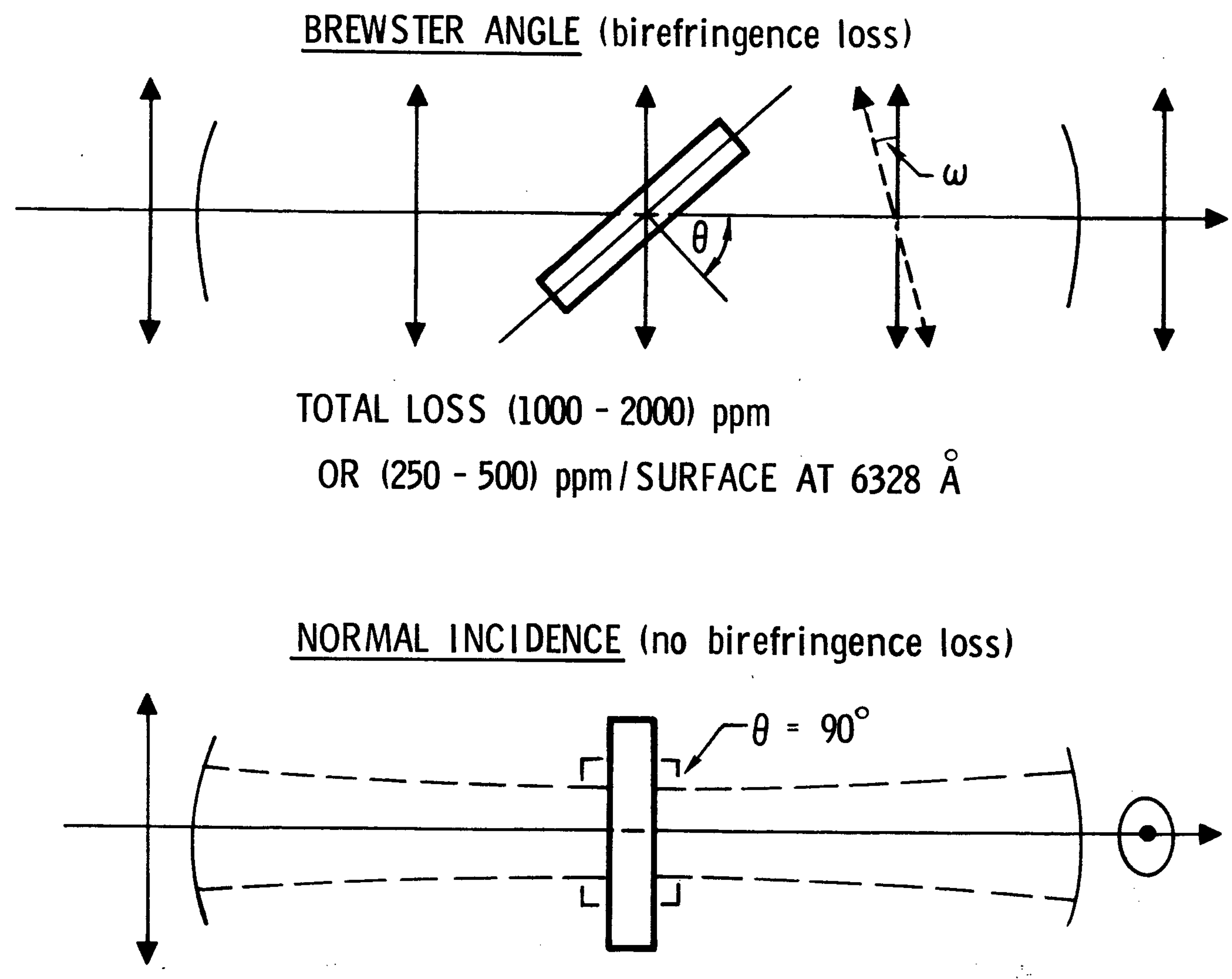

Fig. 4. Two Specific Orientations of Quartz Substrate Sample at Brewster and Normal (Perpendicular) Incidence with Optical Resonator

predicted. Thus, the difference between the two measurements of total loss for these two substrate positions affords a very sensitive measurement of the birefringence of the quartz substrate itself. We were also able to measure the small changes in the birefringences that occur when mechanical stress is applied or removed from the substrate.

By means of the cavity-attenuated phase-shift (CAPS) method, we were able to very quickly establish the best (fastest, easiest, and most repeatable, with the lowest amount of scattering) procedure, which consisted of nothing more than a few light wipes, a few hard wipes, followed by a few more light wipes with ordinary lens paper saturated with the "super solvent."

After cleaning, the substrates were spring loaded into the coating mounting block designed to fit into the commercial coating machines (Fig. 5). This block was made from stainless steel that had been specially cleaned to remove all machining oils and greases. It was then preheated to 300°C for 2 hr to remove any residual contamination. The block was not touched by human hands after this. After being loaded with the cleaned substrates, the block was then placed into a special container that formed a vacuum seal. The container was first evacuated and filled with dry filtered nitrogen, then "hand carried" to the supplier for coating. It was returned the same way. Transportation was not trusted to courier services. If dropped, which had happened, the coated substrates could become jammed into the block and could only be removed with the force of a hammer and wood dowel.

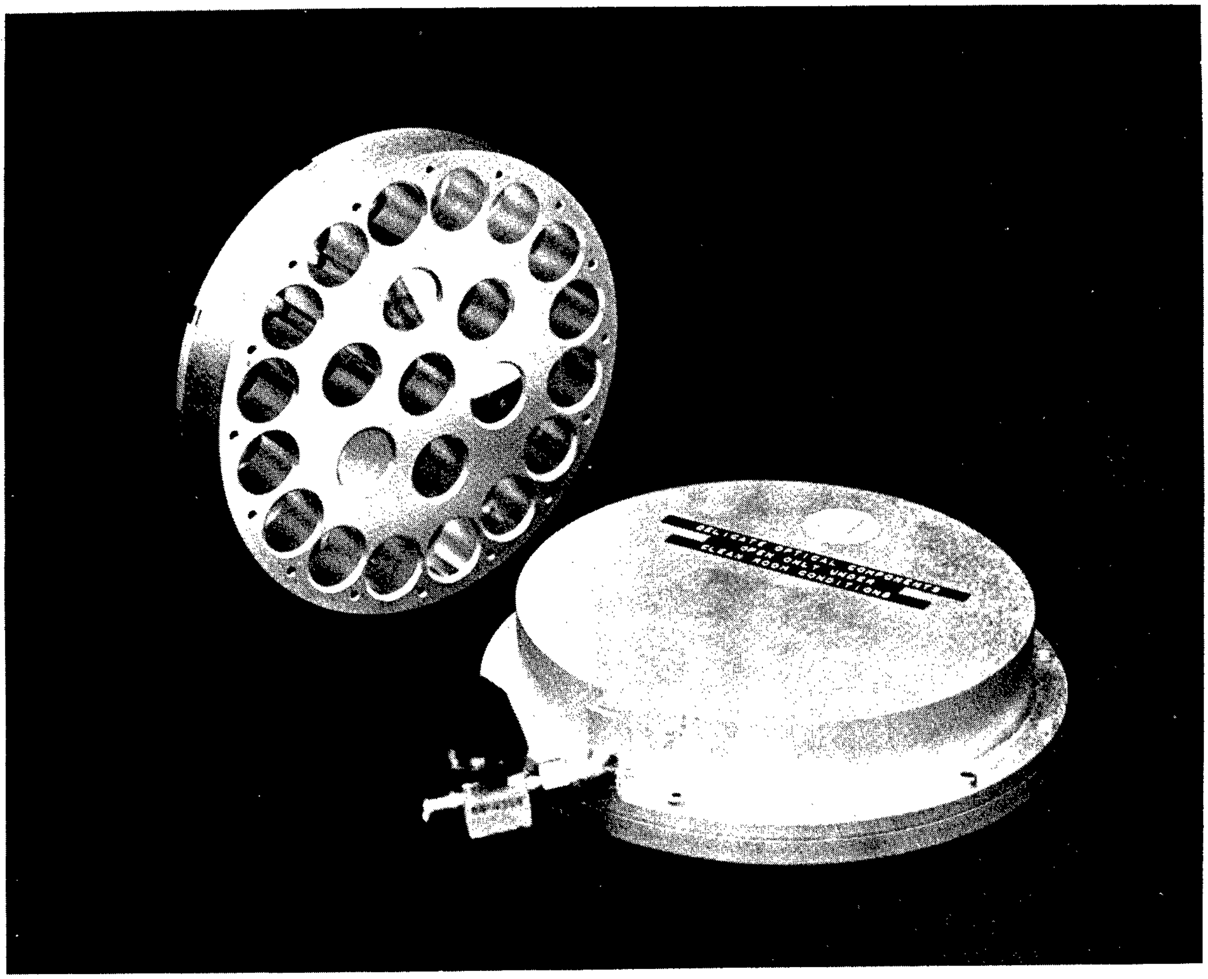

Fig. 5. Mounting Block and Cover Used for Transporting and Coating Cleaned Substrates. The container can be evacuated and filled with an inert gas for storage.

Two commercial vendors were investigated to determine the quality of the optical coatings produced. The first of these was CVI, Albuquerque, New Mexico, and the second vendor was Optical Coating Laboratory Incorporated (OCLI). Our criteria were minimum absorption, scattering and transmission losses, and maximum reflectivity. We decided to test the CVI coating quality with a set coated at the 6328-Å wavelength. Using the CAPS method, we measured a reflectivity of $R = 99.89$, which was surprisingly low. We had expected it to be better, or $R = 99.95$ based upon a stipulated transmission loss of 100 ppm and an anticipated scattering loss of 400 ppm at 6328 Å. This expectation had been based upon information supplied to us by V. Sanders, Litton Industries. We then examined the reflectivity as a function of the incident angle by means of the arrangement shown in Fig. 6. We quickly discovered that, at an angle of $\theta = 45$ deg, the reflectivity approached the anticipated higher value. CVI confirmed our suspicions, i.e., that by mistake, the coating had been given a $\theta = 45$ deg maximum reflectivity coating, which is the usual coating for laser gyro mirrors.

We decided to obtain an OCLI coating at the required wavelength of 8742 Å. The same preparation and handling procedures were again employed, and measurement of the final product yielded a value of $R = 99.975$. The calculations used to predict the reflectivity of the final product, are:

1. For the coating, $R = 1 - (T + A + S)$, $T < 100$ ppm, $A \simeq 30$ to 300 ppm; $S_C \simeq 50$ to 1000 ppm, and $\lambda = 6328$ Å.
2. For the substrate, $S_S \simeq 50$ to 500 ppm.

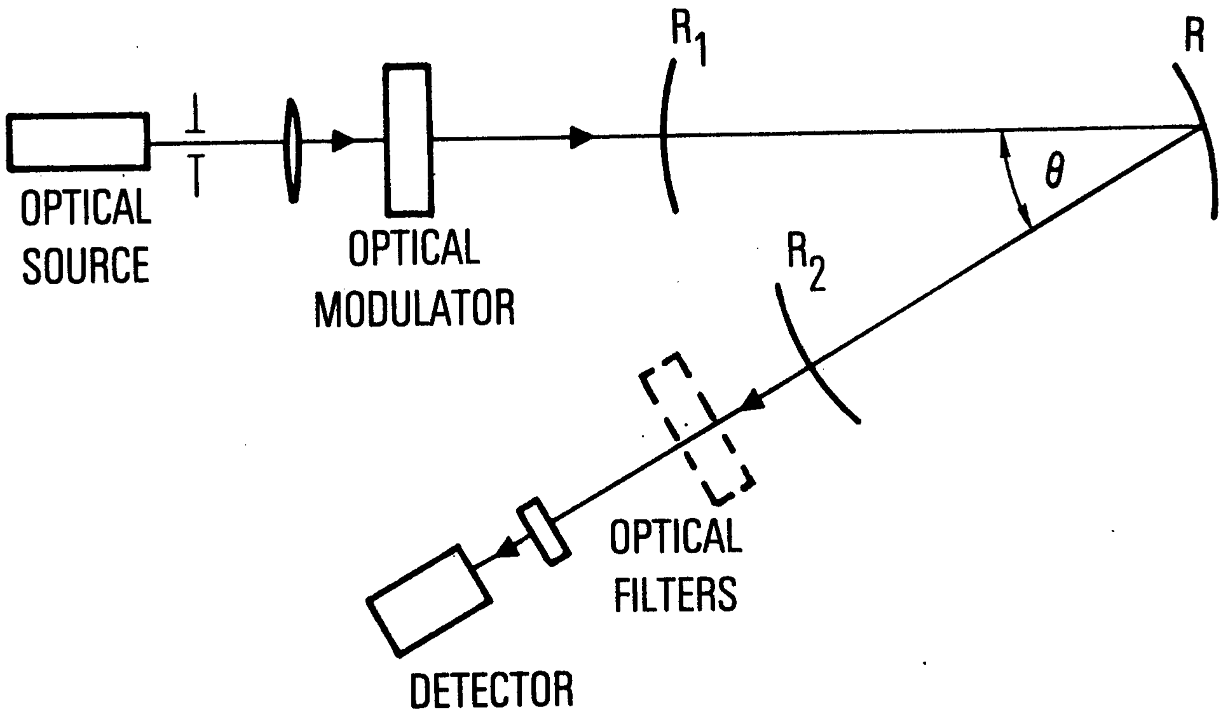

Fig. 6. Alternative Three-Mirror Optical Resonator Configuration for Use with Nontransparent Substrate Mirror Reflectance Measurement at Angle Θ

At $\lambda = 8742$ Å, $S = (6328/8742)^2 \ (S_C + S_S) \ = 0.52\ (50 + 50) = 52$ ppm, $A \leqslant 100$ ppm, $T = 100$ ppm, $R(8742) \geqslant 0.99975$ or $(1 - R = 0.00025)$.

These calculations include the range of values for transmission T, absorption A, and scattering S losses arising from the coating and the substrate at the helium-neon wavelength (6328 Å) for which the most data are available. Wavelength correction of the anticipated scatter from the coating and measured scatter from the substrate is made and combined with the specified absorption and transmission losses to predict the R = 99.975 reflectivity mirrors, which we indeed obtained.

IV. SUMMARY

We found that the use of the cavity-attenuated phase-shift (CAPS) method, in conjunction with simple cleaning practices, results in a simplified procedure for producing very-high-reflectivity mirrors. Our substrate cleaning process is a straightforward procedure. It is reasonable to project that a similar application of this CAPS method to the coating processes itself could permit an order of magnitude improvement over present coating limits. The basic reason is that, unlike other methods, the higher the reflectivity of the mirrors being generated, the more sensitive the phase-shift method becomes. Consequently, there is essentially no limit, other than the materials themselves, in producing extremely high reflectivity mirrors.

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